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TITLE: PLUTONIUM ISOTOPIC MEASUREMENT FOR SMALL PRODUCT SAMPLES

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PLUTONIUM ISOTOPIC MEASUREMENT FOR SMALL PRODUCT SAMPLES*

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Abstract

An automated at-line plutonium isotopic analysis system for small-product plutonium samples in the range of 10 to 1000 μg has been developed. The analysis is based on low-energy gamma rays at 43.48 keV (^{238}Pu), 45.23 keV (^{240}Pu), 51.63 keV (^{239}Pu), 64.83 keV (^{241}Pu-^{237}U), 129.3 keV (^{239}Pu), and 148.6 keV (^{241}Pu). Within a 20-ks counting time, we demonstrated that for plutonium masses >600 μg the precision is <0.7% for ^{238}Pu/^{239}Pu and ^{240}Pu/^{239}Pu ratios and ~4% for the ^{241}Pu/^{239}Pu ratio. The agreement between the results measured by gamma-ray spectroscopy and those measured by mass spectrometry for seven samples is within the gamma-ray precision limits.

1. Introduction

We have developed an automated at-line plutonium isotopic analysis system for nondestructive assay of product samples in the range of 10 to 1000 μg. The system is based on a high-resolution gamma-ray technique, and is designed to provide process control and accountability information for a process line.

Recently, the nondestructive gamma-ray technique has been used to determine plutonium isotopic compositions in bulk samples.[1-3] The technique utilizes either a small planar hyperpure germanium detector (1 or 2 cm^2) to analyze gamma-ray spectra in the 100- to 400-keV range or a large germanium or Ge(Li) photon detector (~70 cm^2) to analyze gamma-ray spectra in the 120- to 640-keV range. In general, it successfully determines the isotopic compositions for ^{238}Pu, ^{239}Pu, and ^{241}Pu within a few hours count time. However, the precision for ^{240}Pu measurements[3] in both the 160- and 642-keV regions was 2 to 4% within a ~14-h count time for a plutonium sample mass >0.25 g. In this case, a counting time over 200 h would be required to assay a small sample with a plutonium mass <1000 μg to obtain a precision better than 3% for ^{240}Pu. This is not practical for monitoring product materials in a process line. For timely plutonium isotopic analysis of small mass samples, we use the high-intensity, low-energy gamma rays at 43.48, 45.23, and 51.63 keV from ^{238}Pu, ^{240}Pu, and ^{239}Pu, respectively. Although these low-energy gamma rays have been used by Gunnink et al.[4] and Cowder et al.[5] for assay of freshly separated solutions, no one has used this energy region for nondestructive assay of moderately aged samples in which the very intense 59.54-keV line from ^{241}Am and/or ^{237}U dominates the spectra. This paper discusses the measurement method and the results obtained by using low-energy gamma rays for nondestructive assay of small plutonium mass samples.

2. Measurement Method

The isotopic ratio, N(m)/N(n), of two isotopes m and n can be determined by measuring their selected gamma rays a and b, respectively.

$$\frac{N(m)}{N(n)} = \frac{R(a)}{R(b)} \cdot \frac{I(b)}{I(a)} \cdot \frac{t(m)}{t(n)} \cdot \frac{\varepsilon(b)}{\varepsilon(a)} , \qquad (1)$$

where

R = measured count rate of gamma rays,
I = absolute branching intensity of gamma rays,
t = half-life of isotope, and
ε = relative efficiency of selected gamma rays, including detector intrinsic efficiency, counting geometry, attenuation, and sample self-attenuation.

The isotopic ratios of ^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{241}Am are determined by using gamma rays at 43.48, 51.63, 45.23, and 59.54 keV, respectively. The ^{241}Pu/^{239}Pu ratios are measured by 64.83 keV/51.63 keV for ^{241}Pu-^{237}U equilibrium samples (>45 days from uranium separation) and by 148.6 keV/129.3 keV for nonequilibrium samples. The isotopic half-lives and the gamma-ray branching intensities are taken from Refs. 6 and 7, respectively.

Relative efficiency (ε) variations arising from sample self-absorption, detector efficiency, and external absorbers are calculated by using known efficiency points from ^{239}Pu gamma rays at energies (E) 38.66, 51.63, 68.72, 129.3, 144.2, 171.3, 195.7, and 203.5 keV. A simple linear ln ε vs ln E interpolation between two relative efficiency points at 38.66 and 51.63 keV is used to calculate the relative efficiencies at 43.48 and 45.23 keV; interpolation between two relative efficiency points at 51.63 and 68.72 keV is used to calculate the relative efficiencies at 59.54 and 64.83 keV. The relative efficiency points at 129.3, 144.2, 171.3, 195.7, and 203.5 keV are fit to a quadratic to determine the relative efficiency at 148.6 keV.

It is well known that the low-energy plutonium gamma rays from aged plutonium samples are difficult to analyze because they are strongly interfered with by the Compton continuum of the 59.54-keV gamma ray from ^{241}Am and/or ^{237}U. To diminish this difficulty, we have carefully selected a detector with the proper combination of resolution, efficiency, and peak-to-Compton ratio at energies below 60 keV. The selected detector is a hyperpure germanium planar type having dimensions of 1000 mm^2 x 15 mm and a resolution (FWHM) of 540 eV at 122 keV. Figure 1 shows a typical low-energy gamma-ray spectrum for a small aged plutonium sample.

The system shown in Fig. 2 also consists of a Canberra Series 85 multichannel analyser (MCA),

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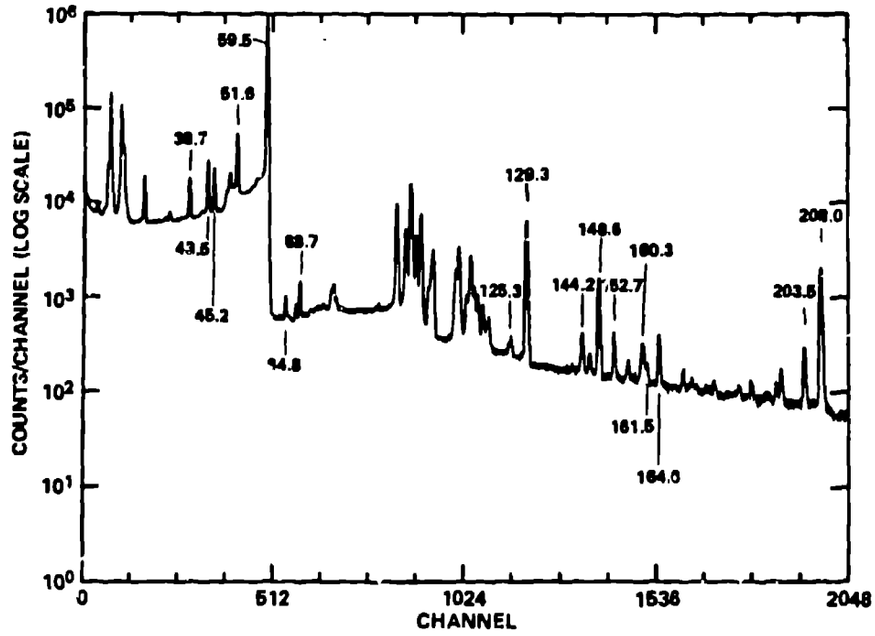

Fig. 1. Low-energy gamma-ray spectroscopy of plutonium.

including an 8-k channel analog-to-digital converter (ADC), and an LSI-11/23 microcomputer and peripherals. The ADC is stabilized by a set of two stabilizers (Canberra 8200) to ensure the long-term stability of the energy calibration of the gamma-ray spectra. The zero and gain stabilization peaks are the 51.63- and 129.3-keV gamma rays from ^{239}Pu. It is expected that no spectral peak will drift more than a few hundredths of a kilovolt from its assumed position, in spite of wide temperature variations at the system location and of wide counting-rate ranges. The MCA is controlled by the LSI-11/23 microcomputer, which has 32-k 16-bit words of memory and is a processor for data acquisition, reduction, and analysis. A Winchester/floppy disk system (DSD-880) provides for storage and transfer of the program and data. The control of assay input and output is accomplished through an LA-120 Decwriter. An automatic data-acquisition-and-analysis program is written in FORTRAN under Digital Equipment Corporation's RT-11 V-4.0 operating system. For setting up a routine assay, the dialogue called up by an operator is kept as simple as possible.

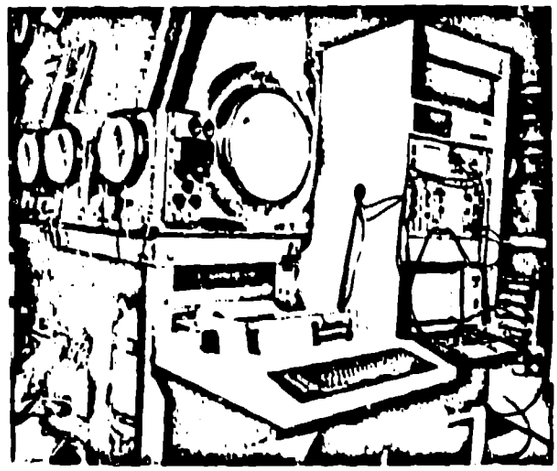

Fig. 2. Photograph of the at-line plutonium isotopic determination system.

3. Results and Discussion

We have analyzed seven aged product samples ranging from 10 to 650 µg of plutonium with americium to plutonium ratios ranging up to 3390 ppm. Table I compares the preliminary gamma-ray spectroscopy results with mass spectrometry results. The uncertainties in the table represent the precision (1σ) of gamma-ray spectroscopy as estimated from counting statistics, including uncertainties from relative efficiencies. Within a 20-ks (~5-1/2 h) counting time, the ^{238}Pu/^{239}Pu and ^{240}Pu/^{239}Pu ratios have a precision ≤0.7% for sample mass >600 µg of plutonium. The precision of ~4% for ^{241}Pu/^{239}Pu is expected to be improved proportionally with the square root of the counting time and sample mass. The agreement between the results measured by gamma-ray spectroscopy and those measured by mass spectrometry for plutonium isotopic ratios is very good and is within gamma-ray precision limits. These results demonstrate that the present plutonium isotopic

TABLE I

COMPARISON OF ISOTOPIC RATIOS BY GAMMA-RAY SPECTROSCOPY WITH MASS SPECTROMETRY

Sample	Approx. Pu Mass (μg)	Am/Pu[c] (ppm)	Ratio: Gamma Spectroscopy[a] / Mass Spectroscopy[b] $^{238}Pu/^{239}Pu$	$^{240}Pu/^{239}Pu$	$^{241}Pu/^{239}Pu$
1	650	306	0.991 ± 0.006	1.001 ± 0.007	0.967 ± 0.040
2	600	372	1.006 ± 0.006	1.001 ± 0.007	0.999 ± 0.042
3	300	1062	-	0.997 ± 0.010	0.939 ± 0.065
4	160	950	-	0.985 ± 0.015	1.128 ± 0.079
5	55	24	1.004 ± 0.211	0.994 ± 0.119	1.078 ± 0.194
6	15	120	-	1.047 ± 0.022	0.935 ± 0.110
7	10	3390	0.974 ± 0.047	1.038 ± 0.054	1.038 ± 0.192

[a]Counting time = 20 ks (~5-1/2 h).
[b]Plutonium-238 determined by radiochemistry.
[c]Determined by gamma-ray spectroscopy.

analysis technique, which is based on high-resolution, nondestructive gamma-ray spectroscopy in the low-energy region, is successful in measuring small product samples in the 10- to 1000-μg range. Both the precision and accuracy of the measurements satisfy the requirements for process control and nuclear safeguards.

4. Acknowledgments

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